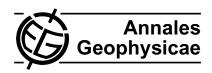
Ann. Geophys., 28, 2149–2159, 2010 www.ann-geophys.net/28/2149/2010/ doi:10.5194/angeo-28-2149-2010 © Author(s) 2010. CC Attribution 3.0 License.



Seasonal stratospheric intrusion of ozone in the upper troposphere over India

S. Fadnavis, T. Chakraborty, and G. Beig

Indian Institute of Tropical Meteorology, Pune, India

Received: 2 July 2010 - Revised: 16 November 2010 - Accepted: 16 November 2010 - Published: 26 November 2010

Abstract. The Model for Ozone and Related chemical Tracers-2 (MOZART-2) is used to examine the evolution of pollutant O_3 in the upper troposphere over the Indian region. Vertical profiles of ozone mixing ratio retrieved from Microwave Limb Sounder (MLS) aboard Earth Observing System (EOS) AURA satellite for the period 2005-2009 and Tropospheric Emission Spectrometer (TES) aboard (EOS) AURA for the period 2006–2007 has been analyzed. The satellite observations reveal the evidence of downward propagation of ozone (100-200 ppb) due to stratospheric intrusion during the winter and pre-monsoon seasons. The regular feature of enhancement of ozone in the upper troposphere over India is presented. Results obtained by the MOZART-2 simulations (for years 2000-2005) confirm the observations and indicate stratospheric intrusion of O₃ during winter and pre-monsoon seasons. Observed enhanced O₃ mixing ratio in the upper troposphere is explained by, variation of Potential Vorticity (PV), tropopause pressure, relative humidity and CO-O₃ correlation.

Keywords. Atmospheric composition and structure (Middle atmosphere – composition and chemistry)

1 Introduction

Upper tropospheric ozone is an important greenhouse gas that affects global outgoing long wave radiations (Worden et al., 2008). Upper tropospheric ozone changes have a great impact on the surface temperature (Forster and Shine, 1997; Intergovernmental Panel on Climate Change (IPCC), 2001, 2007). The increase of tropospheric ozone since preindustrial times has produced a global-averaged radiative



Correspondence to: S. Fadnavis (suvarna@tropmet.res.in)

forcing of $0.35~W~m^{-2}$ (IPCC, 2001). Positive radiative forcing of tropospheric ozone is comparable to that of halocarbons, and twice that of N_2O , but lower than the radiative forcing from CO_2 and CH_4 (IPCC, 2001, 2007). Upper tropospheric ozone enhancement would be the result of convective lifting of surface ozone, in situ production by anthropogenic NO_x , lightning NO_x and partly by HO_x radicals produced from convectively lifted formaldehyde (Cooper et al., 2008) and stratospheric intrusion. Stratosphere to troposphere transport processes are major contributors to the tropospheric ozone budget. Enhanced upper tropospheric ozone can impact air quality if it is transported to the boundary layer (Fiore et al., 2002).

The role of stratosphere-troposphere exchange in the chemical budget of ozone and other species is an important issue. During intrusion most of the trace gases transported from stratosphere spread into surrounding troposphere owing to horizontal mixing. Thus intrusion can reach down to the surface. Downward convective fluxes transport stratospheric/upper tropospheric air into the lower troposphere. An accurate knowledge of transport processes is of great importance in quantifying the roles of transport and chemistry in the budget of trace gases (Brioude et al., 2006). Austin (1991) and Follows (1992) reported that stratospheric intrusions are due to tropopause folding or the strong vertical wind shear. Baray et al. (1998, 2000) discussed the ozone enhancement in the upper troposphere. These studies suggest that subtropical jet stream could be the origin of tropopause folding, leading to stratospheric intrusion to the troposphere. Kim et al. (2002) studied stratospheretroposphere exchange processes over Korea. They reported that ozone intrusion in the upper troposphere corresponds to central axis of the jet stream near the tropopause. The maximum ozone by Stratosphere Troposphere Exchange (STE) occurs in winter-spring time when jet stream was located over Korea. Some of the recent studies reported STE processes over the tropics (Ricaud et al., 2007; Leclair de Bellevue et al., 2007; Das, 2009). These studies are mostly confined to STE process during cyclone or land convection. Over the Indian region, stratosphere troposphere ozone exchange has been reported from ozonesonde, aircraft or mesosphere-stratosphere-troposphere (MST) radar, in station data for particular period (Mitra, 1996; Mandal et al., 1998; Sahu et al., 2009; Gupta et al., 2007; Kumar and Uma, 2009). Impact of transport on distribution of ozone and related trace gases over the Bay of Bengal is reported by Lal et al. (2007). However, seasonal occurrence of STE over India is not reported yet. Intra-decadal variation of stratosphere troposphere exchange of water vapor over the Tibetan plateau is reported by RuiFen and JianPing (2008).

This paper reports seasonal enhanced ozone in the upper troposphere over the Indian region due to stratospheric intrusion. For this purpose, the vertical profiles of ozone mixing ratio obtained from two satellites (1) Microwave Limb Sounder (MLS) for the period 2005–2009, (2) Tropospheric Emission Spectrometer (TES) for the period 2006–2007 over the India region (5–40° N, 65–100° E) are analyzed. We use the global chemistry transport model the MOZART-2 simulations (for the years 2000-2005) to understand transport processes responsible for observed ozone enhancement over the Indian region. Simultaneously CO-O₃ correlation and variation in meteorological parameters such as National Centers for Environmental Prediction (NCEP) tropopause pressure, relative humidity variation and Potential Vorticity (PV) obtained from the European Centre for Medium Range Weather Forecasts (ECMWF) analysis are also examined to explain the observed feature. NCEP determines tropopause from lapse rate. The model used in the NCEP reanalysis has 28 vertical levels extending from the surface to ~40 km, with vertical resolution of \sim 2 km near the tropical tropopause and \sim 1.5 km near the subtropical tropopause. The NCEP output includes global analyses of tropopause temperature and pressure, which are derived from temperature analyses on model sigma levels. The tropopause pressure level is defined by the standard lapse rate criterion; that is, it is identified by the lowest level (above 450 mbar) where the temperature lapse rate becomes less than 2 K km⁻¹; it is not allowed to be higher than 85 mbar. The tropopause pressure is estimated by deriving the lapse rate at each model sigma level and estimating (by interpolation in height) the pressure where the threshold value o f 2 K km⁻¹ is reached. Analyzed temperatures are then interpolated to this level. This algorithm produces tropopause estimates which vary smoothly in space and time (Randel et al., 2000).

2 Model and data analysis

2.1 Model description

The Model for Ozone and Related chemical Tracers-2 (MOZART-2) is a global chemical transport model of at-

mospheric composition designed to simulate tropospheric chemical and transport processes. It has a resolution of \sim 1.8 degrees in longitude and latitude, and includes 31 vertical levels from the surface to the pressure level of approximately 10 mb. This model resolution allows for a description of patterns that extend over typically 200 km, but is too coarse to provide information on smaller scale features. MOZART-2 is driven by assimilated meteorological data corresponding to years 2000–2005 from the European Center for Medium Range Weather Forecasts (ECMWF). The model accounts for the distribution of 63 chemical species subject to 168 reactions including 33 photolysis reactions. The emissions due to fossil fuel combustion, agricultural burning, bio-fuel, etc. are taken from the recent estimates for the Indian region (Beig and Brasseur, 2006) where as rest of the world, it is adopted from the growth pattern based on POET inventory (Granier et al., POET, a database of surface emissions of ozone precursors, 2000-2005, available at http://accent.aero.jussieu.fr/POET.php). Since emission inventory over the Indian region is available until 2005 only, the model simulation presented in the present work corresponds to the years 2000-2005.

2.2 Satellite data sets

2.2.1 Microwave Limb Sounder (MLS) data

The Aura Microwave Limb Sounder (MLS) aboard Earth Observing System (EOS) is one of four instruments on the NASA's EOS Aura satellite, launched on 15 July 2004. The Aura orbit is sun-synchronous at 705 km altitude with 98 inclinations (Waters et al., 2006). EOS MLS version 2, level 2 data are used in the present study. In the UTLS region the MLS V2.2 O₃ data are less sensitive to contamination from cloud effects (Livesey et al., 2007). Vertical resolution is ~4 km in the UTLS region. MLS ozone data quality is good at the altitudes above 215 mb pressure level. For ozone data points having negative 2GPprecision values are not taken into consideration. MLS ozone mixing ratio data are obtained from the website http://disc.gsfc.nasa.gov/data/datapool/MLS.

In the present study MLS ozone data are averaged over the Indian region (5° N to 40° N and 65° E to 100° E). The area averaged ozone are computed for August 2004 to June 2009 between 215–68 mb (215, 146, 100, 68). The daily area averaged mixing ratios are then averaged for a month. Monthly mean profiles are plotted to study seasonal variations within the upper troposphere and lower stratosphere.

2.2.2 Tropospheric Emission Spectrometer (TES) data

The Tropospheric Emission Spectrometer (TES) launched into sun-synchronous orbit aboard Aura, the third of NASA's Earth Observing System (EOS) spacecraft, on 15 July 2004. TES provides a global view of tropospheric trace gas profiles

including ozone, water vapor, and carbon monoxide, along with atmospheric temperature, surface temperature and emissivity, effective cloud top pressure, and effective cloud optical depth (Worden et al., 2004). For cloud free conditions, the vertical resolution of TES ozone profile retrievals is typically 6 km in the tropics (Jourdain et al., 2007; Worden et al., 2004). TES measurements are available since 2005. TES Level 2 data with grid spacing 4° longitude × 2° latitude is analyzed in the present study. Tropospheric ozone retrievals from TES have been validated against ozonesonde and lidar measurements and it is generally found that the values are biased high by as much as 15% after accounting for the TES vertical resolution and a priori constraint (Worden et al., 2009). TES monthly mean ozone mixing ratios are obtained from the website: ftp://l4ftl01.larc.nasa.gov/TES/TL2O3N.

The TES data is not available for some months during 2005 and 2008; hence the analysis presented here uses TES data during the years 2006 and 2007. Monthly mean profiles are plotted to study seasonal variations within the troposphere and the lower stratosphere.

The horizontal and vertical distributions of ozone are analyzed using TES and MOZART model simulations. Relative Humidity (RH) and tropopause pressure are obtained from NCEP/NCAR reanalysis data. Potential vorticity is obtained from ECMWF analysis. The potential vorticity is expressed in PV units ($\rm m^2 \, s^{-1} \, K \, kg^{-1}$). One potential vorticity unit (1 PVU) is taken as $1.0 \times 10^{-6} \, \rm m^2 \, s^{-1} \, K \, kg^{-1}$. Air with potential vorticity of 2 PVU or more is taken to be of stratospheric origin (Holton et al., 1995; Newell et al., 1997). In the discussion that follows, seasons are considered as pre monsoon (March-April-May), summer monsoon (June-July-August-September) and winter (December-January-February).

3 Results and discussions

Figure 1a exhibits vertical distributions of monthly mean ozone mixing ratios (averaged over the Indian region) as obtained from MLS satellite data during August 2004 to June 2009. The thick black line with dots indicates the tropopause pressure. In the upper troposphere and lower stratosphere ozone mixing ratio varies from 100-800 ppb. The most evident feature is that every year there are enhanced values of ozone in the upper troposphere during February-March-April. During majority of years there is ozone enhancement in the upper troposphere with mixing ratios \sim 200 ppb during February. Ozone mixing ratio ~100 ppb is observed near 200 mb during the months of February-March-April of every year. During winter season tropopause is at lower altitudes (tropopause pressure \sim 120–130 mb). Low tropopause pressure forms favorable condition for STE process (Das et al., 2009). Hence stratospheric ozone mixing ratio may cross tropopause and enter upper troposphere.

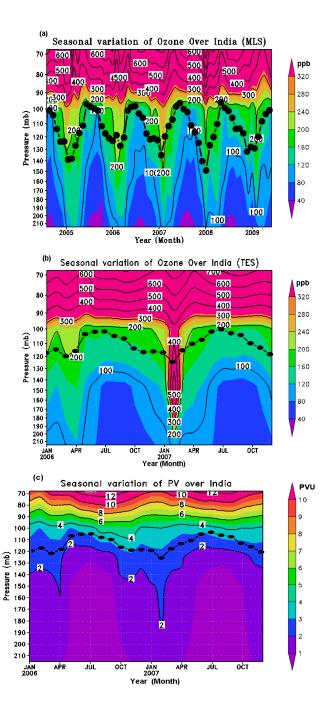


Fig. 1. (a) Vertical variation of monthly mean ozone mixing ratio (averaged over the Indian region) in the upper troposphere as obtained from MLS satellite data during August 2004 to June 2009. (b) Vertical variation of ozone mixing ratio (averaged over the Indian region) in the upper troposphere as obtained from TES satellite measurements during 2006–2007. (c) Vertical variation of potential vorticity (averaged over the Indian region) in the upper troposphere as obtained from ECMWF analyses during 2006–2007. Thick black line with dots indicates tropopause pressure.

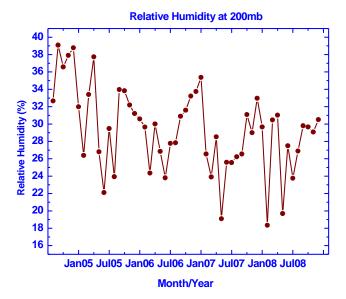


Fig. 2. Variation of relative humidity obtained from NCEP/NCAR reanalysis data at 200 mb during August 2004–June 2009.

Figure 1b shows seasonal variation of ozone in the upper troposphere as obtained from TES satellite data during 2006–2007. Thick black line with dots indicates tropopause pressure variation. Similar to MLS satellite observations, TES data also shows enhanced ozone mixing ratios ~200 ppb in the upper troposphere during every February. Ozone mixing ratios are seems to be propagating downward. It may crosses the tropopause and reaches to 160–180 mb pressure levels. Ozone mixing ratios ~100 ppb are observed near 200 mb during February-March-April. It should be noted that both MLS and TES observations show greater ozone enhancement in the upper troposphere (may be due to intrusion) during the month of February during 2006–2007.

Figure 1c depicts vertical variation of potential vorticity obtained from ECMWF analysis (averaged over the Indian region) during 2006-2007 with tropopause pressure over plotted (The thick black line with dots). It indicates that PV more that 2 PVU is observed near the upper troposphere throughout the year. But, most noticeable feature is that, tongue of air with PV more than 2 PVU penetrates into the upper troposphere up to 160-180 mb pressure levels during every winter and pre-monsoon seasons. During 2007 it penetrates into the upper troposphere near the pressure level \sim 145 mb during post-monsoon season too. These pressure levels coincide with observed enhanced ozone mixing ratios in the upper troposphere during same seasons. It is interesting to compare vertical variation of PV (Fig. 1c)and ozone mixing ratio (Fig. 1b) during 2006 and 2007. PV equal to 2 PVU penetrates into the upper troposphere and reached to 160 mb during April 2006 and 180 mb during February 2007. Enhanced ozone mixing ratio \geq 200 ppb is observed at these pressure levels during same the period. This indicates that ozone mixing ratios at upper tropospheric pressure levels has its origin in the stratosphere.

Figure 2 indicates variation of NECP derived Relative Humidity (RH) at 200 mb. At this pressure level RH varies between 16%–38%. During February-March-April, especially during February RH near 200 mb is very low (< 25%). Enhanced ozone mixing ratio in correlation with very low RH also indicates dry air arrived near 200 mb may be of stratospheric origin. Over Europe, Tulet et al. (2002) also observed ozone mixing ratio \sim 110 ppb between 500–300 mb with low RH (20-40%) during stratospheric intrusion. Hence, observed enhanced ozone mixing ratio (\sim 100 ppb) near 200 mb level may be due to stratospheric intrusion. To identify stratospheric intrusion across the tropical tropopause, we also used the correlation between CO and ozone. Waugh et al. (1997) introduced the concept for tracers (N2O versus NO_v) correlation to investigate tropospheric transport. They used the concept of mixing lines to identify cross tropopause mixing. Hoor et al. (2002) and Hipskind et al. (1987) used the correlation between CO and ozone to identify exchange across extra-tropical tropopause. Hoor et al. (2002) explained that if we neglect mixing processes across the tropopause, the idealized relationship between ozone and CO can be expected to display L-shape (Fischer et al., 2000). The nearly constant stratospheric equilibrium values of CO at strongly increasing ozone gives rise to a stratospheric branch, whereas the tropospheric branch is characterized by low ozone but highly variable CO values ranging from 50-200 ppb. In order to confirm the stratospheric intrusion correlation is studied from TES satellite observed CO and O₃ at upper tropospheric pressure levels during February-April 2007. Correlation plots are obtained from time series of monthly mean ozone and CO mixing ratio averaged over the Indian region. Figure 3a-j indicates TES ozone and CO mixing ratio correlation plot at 215, 146, and 100 mb level during February, March and April 2007, respectively. The L-shape curves at these pressure levels show high ozone $(O_3 > 350 \text{ ppb})$ mixing ratios indicate stratospheric branch. Strong negative correlation varying between -0.93and -0.87 is observed at upper tropospheric pressure levels. The opposite gradients of the two species indicate the ozone toward the stratospheric air and the carbon monoxide toward the tropospheric air. During tropopause fold event Hipskind et al. (1987) also reported negative correlation between O₃ and CO (ranging between -0.98 and -0.54).

To examine transport path ways, we use MOZART-II simulations for the years 2000–2005. Figure 4a–f shows seasonal variation of ozone mixing ratios between 1000 mb to 68 mb as obtained from MOZART model simulations for years 2005–2000, respectively. Thick black color dotted line indicates tropopause pressure. The model simulations show an ozone seasonal cycle which is consistent with the observed intrusion of stratospheric air into the upper troposphere at the beginning of the year. Every year there is strong ozone enhancement in the upper troposphere during

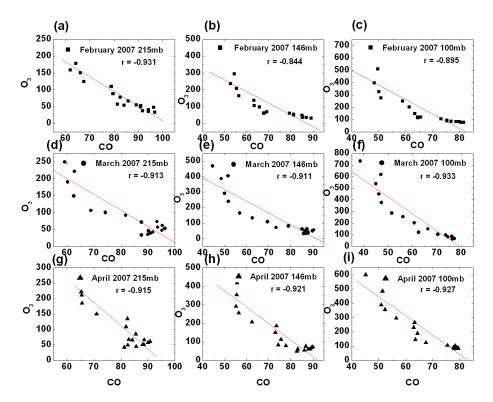


Fig. 3. Correlation between TES ozone and CO volume mixing ratio during (a) February 2007 at 215 mb, (b) February 2007 at 146 mb, (c) February 2007 at 100 mb, (d) March 2007 at 215 mb, (e) March 2007 at 146 mb, (f) March 2007 at 100 mb, (g) April 2007 at 215 mb, (h) April 2007 at 146 mb, (i) April 2007 at 100 mb.

winter and pre-monsoon seasons (except during 2005; where intrusion may be weaker). Ozone mixing ratio $\sim\!200\,\mathrm{ppb}$ crosses the tropopause and enters the upper troposphere during February–April as observed in MLS and TES data. During winter and pre-monsoon seasons of the years 2004–2000, ozone mixing ratio $\sim\!100\,\mathrm{ppb}$ reaches to 300 mb. Thus model simulations also show downward transport of ozone mixing ratios during winter and pre-monsoon seasons as observed in satellite measurements.

Since ozone intrusion mainly occurs during February-March-April months, ozone mixing ratios are averaged over these months at every pressure level. These seasonally averaged ozone mixing ratios are then averaged over the Indian longitudes (65-100° E). The latitude-altitude distribution of zonal and seasonal (February-March-April) averaged ozone mixing ratios as obtained from TES measurements for the year 2007 and model simulation for the years 2002-2005 is shown in Fig. 5a-e. These figures indicate that ozone intrusion occur between 20-40° N latitudes. Ozone mixing ratio \sim 400 ppb reaches up to 170 mb and 200 ppb near 300 mb at 35° N. Between the latitudes 20° N and 30° N ozone mixing ratio \sim 100–200 ppb reaches to 215 mb pressure level. Similar results are observed in model simulation of the years 2001-2000 (figures not included). Latitude-altitude cross section of ozone mixing ratio variation during 2005 (Fig. 5b) shows that ozone mixing ratio \sim 300–400 ppb enters the troposphere (at the altitudes below 100 mb) over the narrow region 32–40° N. This may be the reason that seasonal variation of ozone show weaker intrusion (as observed in Fig. 4a) during 2005. Figure 5f shows latitude-altitude cross-section of PV. PV more than 2 PVU is observed between latitudes 20° N and 40° N, at the pressure levels above $100{-}120$ mb. This confirms that the ozone intrusion might have occurred between 20° N and 40° N as seen in observational data and model simulated results.

Latitude-longitude cross-section of ozone distribution (averaged for February-March-April months) as obtained from TES observations and model simulations is examined at every upper tropospheric pressure level between 216 mb and 100 mb. Figure 6a-d depicts the horizontal distribution of seasonally averaged (February-March-April) ozone at 146 mb and 215 mb pressure levels as obtained from TES observations for the year 2007 (Fig. 6a-b) and model simulations for the year 2005 (Fig. 6c-d). Figure 6a shows that enhanced ozone mixing ratio is observed in the region with latitudes 30–36° N and longitudes 85–95° E. Ozone intrusion at 146 mb level may be occurring keeping center within this region. Ozone mixing ratio \sim 600 ppb is observed in the location near the point 33° N, 93° E. In the region between the latitudes 30–40° N (over all the longitudes) ozone mixing ratios varies between 200–600 ppb. Similar structure of ozone distribution (but enhanced ozone mixing ratio) is observed

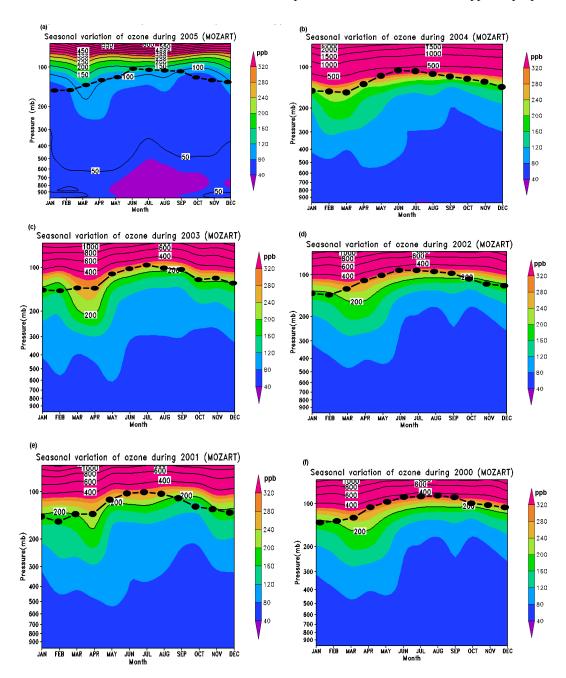


Fig. 4. Vertical variation of ozone mixing ratio (averaged over the Indian region) as obtained from analysis of MOZART model simulations for the years (a) 2005, (b) 2004, (c) 2003, (d) 2002, (e) 2001, (f) 2000. Thick black line with dots indicates tropopause pressure.

at 100 mb (figure not shown). Figure 6b exhibits the horizontal distribution of ozone at 215 mb pressure level. Enhanced ozone mixing ratio $\sim\!200\text{--}300\,\mathrm{ppb}$ is observed in the region with latitudes 30–36° N and longitudes 85–95° E. At 316 mb pressure level (figure not shown) ozone mixing ratio $\sim\!100\,\mathrm{ppb}$ is observed in the same region (latitudes 30–36° N and longitudes 85–95° E) and 70–100 ppb between 30–40° N (over all the longitudes). Figure 6c and d depicts ozone distribution (seasonally averaged) at 146 mb and 215 mb pres-

sure levels as obtained from model simulation for the year 2005. MOZART model simulation does not indicate very high ozone mixing ratio in the region with latitudes 30–36° N and longitudes 85–95° E as observed in TES satellite data. This may be due to coarse model resolution \sim 1.8 degrees in longitude and latitude. At 146mb, ozone mixing ratio \sim 150–400 ppb (Fig. 6c) and at 215 mb ozone mixing ratio \sim 100–150 ppb are observed between the latitudes 30–40° N which is in agreement with TES observations. Figure 6e

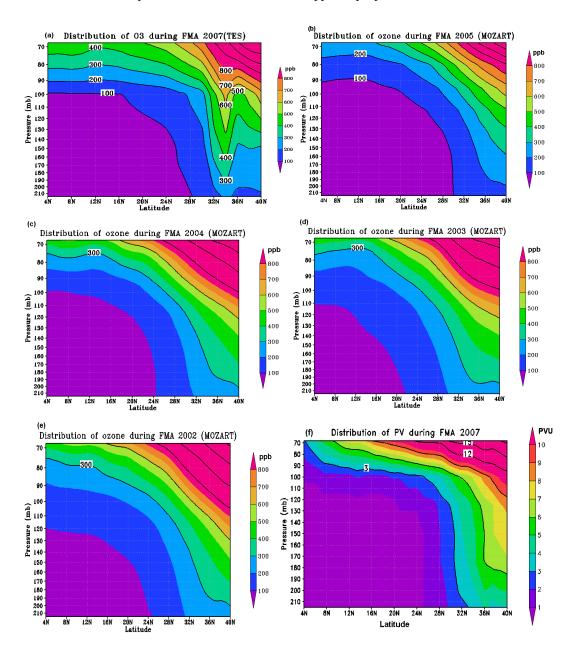


Fig. 5. Latitude-height cross section of zonal (averaged over the longitudes 65–100° E) and seasonal (February-March-April months) mean ozone mixing ratio as obtained from (a) analysis of TES observations for the year 2007; (b) analysis of MOZART model simulation for the year 2005; (c) analysis of MOZART model simulation for the year 2004; (d) analysis of MOZART model simulation for the year 2003; (e) analysis of MOZART model simulation for the year 2002; (f) latitude-height cross section of zonal (averaged over the longitudes 65–100° E) and seasonal (February-March-April months) mean potential vorticity for the year 2007.

and f exhibits spatial distribution of zonal (longitudes 65° E–100° E) and seasonal (FMA) averaged potential vorticity at 146 mb and 215 mb pressure levels respectively. PV more than 2 PVU is observed between the latitudes 28° N and 40° N at 146 mb and 215 mb pressure levels. This indicates that enhance ozone mixing ratios observed from TES measurements and model simulation at these latitudes and altitudes are of stratospheric origin. From ozonesonde observations Kim et al. (2002) showed that stratospheric ozone is

transported down to the upper troposphere by strong zonal wind speed of upper level jet stream. The enhancement of ozone mixing ratio in the troposphere occurs during winter spring time over Korea. It should be noted that during winter location of East Asian Subtropical Westerly Jet (EASWJ) near 200 mb is generally along 28° N, 90° E and pre-monsoon season (April–May) it jumps to nearby 32° N and migrates northward to 40° N (Zhang et al., 2008). Thus enhanced ozone mixing ratios in the upper troposphere as

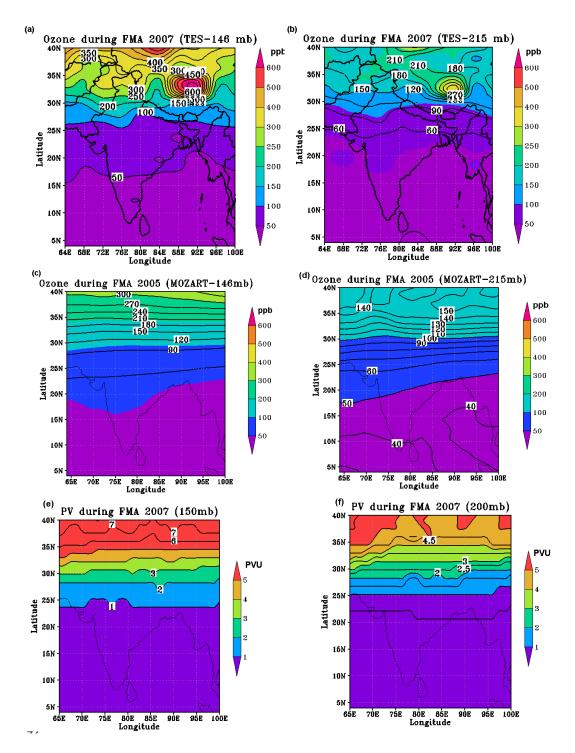


Fig. 6. Latitude-longitude distribution of seasonal averaged ozone (averaged for February-March-April months) over the Indian region (a) at 146 mb as obtained from TES measurements for the year 2007; (b) at 215 mb as obtained from TES measurements for the year 2007; (c) at 146 mb as obtained from MOZART model simulation for the year 2005; (d) at 215 mb as obtained from MOZART model simulation for the year 2005; (e) latitude-longitude cross-section of seasonal averaged (February-March-April) potential vorticity at 150 mb; (f) latitude-longitude cross-section of seasonal averaged (February-March-April) potential vorticity at 200 mb.

observed in satellite data (over the latitudes $20\text{--}40^\circ\,\text{N}$) and model simulations during winter and pre-monsoon seasons may be due to downward transport of ozone from stratosphere due to strong zonal wind speed of the EASWJ. It should be noted that the intense thunderstorm activity during pre-monsoon season also contributes in photochemical production of ozone (via production of NO_x) in the upper troposphere.

4 Discussions on transport mechanism

MLS and TES observations show the evidence of enhanced ozone mixing ratios in the upper troposphere due to downward propagation of ozone mixing ratio (\sim 200 ppb) from the lower stratosphere to the upper troposphere (up to 200 mb) during winter and pre-monsoon seasons when tropopause is at lower altitudes. PV shows PV \geq 2 PVU during these seasons. This indicates tongue of stratospheric air enters the upper troposphere during winter and pre-monsoon seasons. Low relative humidity (<25%) near 200 mb confirms that air mass near 215–100 mb pressure levels is of stratospheric origin. Stratospheric intrusion is also identified using CO-O₃ correlation. Strong negative correlation coefficients and L-shape of CO-O₃ correlation plots at the upper tropospheric pressure levels confirm the stratospheric intrusion.

Latitude-height cross section shows that ozone intrusion occurs between the latitudes 20° N and 40° N. Horizontal distribution of TES ozone mixing ratio (seasonally averaged during February-March-April 2007) indicates that center of intrusion might have been over Indian subtropical region (latitudes 30–36° N and longitudes 85–95° E) during 2007. MOZART model simulation has also reproduced ozone intrusion over the Indian subtropical region during winter and pre-monsoon seasons. Latitude-altitude and latitude-longitude cross-sections of PV show PV > 2 PVU between 20° N and 40° N during these seasons. During winter and pre-monsoon seasons, East Asian subtropical westerly jet (near 200 mb) blow near these latitudes. Downward ozone transport by the strong zonal wind speed of the jet stream may be responsible for stratospheric intrusion. From Tropospheric Emission Spectrometer (TES) measurements, Worden et al. (2009) also observed enhanced ozone mixing (\sim 200 ppb) near 300 mb related to stratospheric intrusion during pre-monsoon season over the Indian region. Sprenger and Wernli (2003) examined 15 years of ERA 15 meteorological data from ECMWF and found stratosphere to troposphere exchange is maximum over central Asia in the premonsoon season.

Hence it is proposed that downward propagation of ozone from the lower stratosphere into the upper troposphere during winter pre-monsoon seasons contributes to ozone enhancement in the upper troposphere over the Indian region. Downward ozone transport by the strong zonal wind speed of the jet stream may be responsible for stratospheric intrusion.

5 Conclusions

The vertical profiles of ozone mixing ratio retrieved from Microwave Limb Sounder (MLS) during 2004–2009 and Tropospheric Emission Spectrometer (TES) for the period 2006-2007 show evidence of enhanced ozone mixing ratio in the upper troposphere during winter and pre-monsoon. Seasonal distribution of potential vorticity shows PV greater than 2 PVU in the upper troposphere during these seasons. This indicates that observed enhanced ozone mixing ratios in the upper troposphere may be due to downward propagation of O₃ (100–200 ppb) from the lower stratosphere to the upper troposphere. Simultaneously very low RH (>25%) and strong negative CO/O₃ correlation (in the upper troposphere) confirms that observed enhanced ozone in the upper troposphere may be due to stratospheric intrusion during winter and pre-monsoon seasons. MOZART model simulations (for the years 2000–2005) confirmed these results.

Seasonal mean latitude-height and latitude-longitude cross sections (at the upper tropospheric pressure levels) of satellite observed ozone and model simulations show that stratospheric intrusion occurs over the Indian subtropical region (25–40° N). Seasonal mean latitude-height and latitude-longitude cross-sections of potential vorticity show PV more than 2PVU over the latitudes 25–40° N. During winter and pre-monsoon seasons, East Asian south westerly jet (near 200 mb) blows over the Indian subtropical region. Downward transport of ozone by the strong zonal wind speed of the jet stream may be responsible for observed seasonal stratospheric intrusion.

Acknowledgements. We acknowledge with gratitude B. N. Goswamy, Director IITM for his encouragement during the course of this study. The authors also thank to anonymous reviewer for their critical comments and valuable suggestions, which helped in improving the scientific value of this research paper.

Topical Editor C. Jacobi thanks J.-L. Baray and another anonymous referee for their help in evaluating this paper.

References

Austin, J. F. and Follows, M. J.: The ozone record at Payerne: an assessment of the cross-tropopause flux, Atmos. Environ., 25A, 1873–1880, 1991.

Baray, J., Baray, L., Ancellet, G., Taupin, F. G., Bessafi, M., Baldy, S., and Keckhut, P.: Subtropical tropopause break as a possible stratospheric source of ozone in the tropical troposphere, J. Atmos. Solar Terr. Phys., 60(1), 27–36, 1998.

Baray, J.-L., Daniel, V., Ancellet, G., and Legras, B.: Planetary-scale tropopause folds in the southern subtropics, Geophys. Res. Lett., 27(3), 353–356, doi:10.1029/1999GL010788, 2000.

Beig, G. and Brasseur, G. P.: Influence of anthropogenic emissions on tropospheric ozone and its precursors over the Indian tropical region during a Monsoon, Geophys. Res. Lett., 33, L07808, doi:10.1029/2005GL024949, 2006.

- Brioude, J., Cammas, J.-P., and Cooper, O. R.: Stratosphere-troposphere exchange in a summertime extratropical low: analysis, Atmos. Chem. Phys., 6, 2337–2353, doi:10.5194/acp-6-2337-2006, 2006.
- Cooper, O. R.: Evidence for a recurring eastern North America upper tropospheric ozone maximum during summer, J. Geophys. Res., 112, D23304, doi:10.1029/2007JD008710, 2007.
- Das, S. S.: A new perspective on MST radar observations of stratospheric intrusions into-troposphere associated with tropical cyclone, Geophys. Res. Lett., 36, L15821, doi:10.1029/2009GL039184, 2009.
- Fiore, A. M., Jacob, D. J., Bey, I., Yantosca, R. M., Field, B. D., Fusco, A. C., and Wilkinson, J. G.: Background ozone over the United States in summer: Origin, trend, and contribution to pollution episodes, J. Geophys. Res., 107(D15), 4275, doi:10.1029/2001JD000982, 2002.
- Fischer, H., Wienhold, F. G., Hoor, P., Bujok, O., Schiller, C., Siegmund, P., Ambaum, M., Scheeren, H. A., and Lelieveld, J.: Tracer correlations in the northern high latitude lowermost stratosphere: Influence of cross-tropopause mass exchange, Geophys. Res. Lett., 27, 97–100, 2000.
- Follows, M. J.: Notes and correspondence; on the cross-tropopause exchange of air, J. Atmos. Sci., 49, 879–882, 1992.
- Follows, M. J. and Austin, J. F.: A zonal average model of the stratospheric contribution to the tropospheric ozone budget, J. Geophys. Res., 97, 18047–18060, 1992.
- Forster, P. M. de F. and Shine, K. P.: Radiative forcing and temperature trends from stratospheric ozone changes, J. Geophys. Res., 102, 10841–10857, 1997.
- Granier, C., Guenther, A., Lamarque, J. F., Mieville, A., Muller, J. F., Olivier, J., Orlando, J., Peters, J., Petron, G., Tyndall, G., and Wallens, S.: POET, a database of surface emissions of ozone precursors, available from http://accent.aero.jussieu.fr/POET.php, last access November 2010.
- Gupta, S., Lal, S., Venkataramani, S., Rajesh, T. A., and Acharya, Y. B.: Variability in the vertical distribution of ozone over a subtropical site in India during a winter month, J. Atmos. Solar Terr. Phys., 69, 1502–1512, 2007.
- Hipskind, R. S., Gregory, G. L., Sachse, G. W., Hill, G. F., and Danielsen, E. F.: Correlations between ozone and carbon monoxide in the lower stratosphere, folded tropopause, and maritime troposphere, J. Geophys. Res., 92, 2121–2130, 1987.
- Holton, J. R., Haynes, P. H., McIntyre, M. E., Douglass, A. R., Rood, R. B., and Pfister, L.: Stratosphere-troposphere exchange, Rev. Geophys., 33(4), 403–439, doi:10.1029/95RG02097, 1995.
- Hoor, P., Fischer, H., Lange, L., Lelieveld, J., and Brunner, D.: Seasonal variations of mixing layer in the lowermost stratosphere as identified by the CO-O3 correlation from in situ measurements, J. Geophys. Res., 107D(5), 4044, doi:10.1029/2000JD000289, 2002.
- Intergovernmental Panel on Climate Change, Climate Change 2001:
 The Scientific Basis Contribution of Working Group I to the
 Third Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Houghton, J. T., Ding, Y., Griggs, D. J.,
 Noguer, M., van der Linden, P. J., Dai, X., Maskell, K., and Johnson, C. A., 881 pp., Cambridge Univ. Press, New York, 2001.
- Intergovernmental Panel on Climate Change, Climate Change 2007: The Physical Science Basis, Summary for Policymakers – Contribution of Working Group I to the Fourth Assessment Report

- of the Intergovernmental Panel on Climate Change, Cambridge Univ. Press, New York, 2007.
- Jourdain, L., Worden, H. M., Worden, J. R., Bowman, K., Li, Q., Eldering, A., Kulawik, S. S., Osterman, G., Boersma, K. F., Fisher, B., Rinsland, C. P., Beer, R., and Gunson, M.: Tropospheric vertical distribution of tropical Atlantic ozone observed by TES during the northern African biomass burning season, Geophys. Res. Lett., 34, L04810, doi:10.1029/2006GL028284, 2007.
- Kim, Y. K., Lee, H. W., Park, J. K., and Moon, Y. S.: The stratosphere-troposphere exchange of ozone and aerosols over Korea, Atmos. Environ., 36A, 449–436, 2002.
- Kumar, K. K. and Uma, K. N.: High temporal resolution VHF radar observations of stratospheric air intrusions in to the upper troposphere during the passage of a mesoscale convective system over Gadanki (13.5° N, 79.2° E), Atmos. Chem. Phys. Discuss., 9, 13843–13857, doi:10.5194/acpd-9-13843-2009, 2009.
- Lal, S., Sahu, L. K., and Venkataramani, S.: Impact of transport from the surrounding continental regions on the distributions of ozone and related trace gases over the Bay of Bengal during February 2003, J. Geophys. Res., 112, D14302, doi:10.1029/2006JD008023, 2007.
- Leclair De Bellevue, J., Baray, J. L., Baldy, S., Ancellet, G., Diab, R., and Ravetta, F.: Simulations of stratospheric to tropospheric transport during the tropical cyclone Marlene event, Atmos. Environ., 41, 6510–6526, 2007.
- Livesey, N. J.: Version 2.2 level 2 data quality and description document, report, NASA Goddard Earth Sci. Distribution Active Arch. Cent., Pasadena, California, 2007.
- Mandal, T. K., Cho, J. Y. N., Rao, P. B., Jain, A. R., Peshin, S. K., Srivastava, S. K., Bohra, A. K., and Mitra, A. P.: Stratospheretroposphere ozone exchange observed with the Indian MST radar and a simultaneous balloon-borne ozonesonde, Rad. Sci., 33(4), 861–893, doi:10.1029/97RS03553, 1998.
- Mitra, A. P.: Troposphere-stratosphere coupling and exchange at low latitude Adv. Space Res., 17(11), 89–97, 1996.
- Newell, R. E., Browell, E. V., Davis, D. D., and Liu, S. C.: Western Pacific tropospheric ozone and potential vorticity: Implications for Asian pollution, Geophys. Res. Lett., 24(22), 2733–2736, doi:10.1029/97GL02799, 1997.
- Randel, W. J., Wu, F., and Gaffen, D. J.: Interannual variability of the tropical tropopause derived from radiosonde data and NCEP reanalyses, J. Geophys. Res., 105(D12), 15509–15523, 2000.
- Ricaud, P., Barret, B., Attié, J.-L., Motte, E., Le Flochmoën, E., Teyssèdre, H., Peuch, V.-H., Livesey, N., Lambert, A., and Pommereau, J.-P.: Impact of land convection on tropospherestratosphere exchange in the tropics, Atmos. Chem. Phys., 7, 5639–5657, doi:10.5194/acp-7-5639-2007, 2007.
- RuiFen, Z. and Li JianPing: Influence of atmospheric heat sources over the Tibetan Plateau and the tropical western North Pacific on the inter-decadal variations of the stratosphere troposphere exchange of water vapor, Science in China Series D-Earth Sciences, 51(8), 1179–1193, 2008.
- Sahu, L. K., Lal, S., Thouret, V., and Smit, H. G.: Seasonality of tropospheric ozone and water vapor over Delhi, India: a study based on MOZAIC measurement data, J. Atmos. Chem., 62(2), 151–174, 2009.
- Sprenger, M. and Wernli, H.: A Northern Hemispheric climatology of cross-tropopause exchange for the ERA15 time period (1979–1993), J. Geophys. Res., 108, 8521, doi:10.1029/2002JD002636,

2003.

- Tulet, P., Suhre, K., Mari, C., Solmon, F., and Rosset, R.: Mixing of Boundary layer and upper tropospheric ozone during a deep convective event over Western Europe, Atmos. Environ., 36, 4491– 4501, 2002.
- Waters, J. W., Froidevaux, L., Harwood, R. S., Jarnot, R. F., Pickett, H. M., Read, W. G., Siegel, P. H., Cofield, R. E., Filipiak, M. J., Flower, D. A., Holden, J. R., Lau, G. K., Livesey, N. J., Manney, G. L., Pumphrey, H. C., Santee, M. L., Wu, D. L., Cuddy, D. T., Lay, R. R., Loo, M. S., Perun, V. S., Schwartz, M. J., Stek, P. C., Thurstans, R. P., Boyles, M. A., Chandra, S., Chavez, M. C., Chen, G.-S., Chudasama, B. V., Dodge, R., Fuller, R. A., Girard, M. A., Jiang, J. H., Jiang, Y., Knosp, B. W., LaBelle, R. C., Lam, J. C., Lee, K. A., Miller, D., Oswald, J. E., Patel, N. C., Pukala, D. M., Quintero, O., Scaff, D. M., Snyder, W. V., Tope, M. C., Wagner, P. A., and Walch, M. J.: The Earth Observing System Microwave Limb Sounder (EOS MLS) on the Aura satellite, IEEE Trans. Geosci. Rem. Sens., 44, 5, 2006.
- Waugh, D. W., Hall, T. M., Randel, W. J., Rasch, P. J., Boville, B. A., Boering, K. A., Wofsy, S. C., Daube, B. C., Elkins, J. W., Fahey, D. W., Dutton, G. S., Volk, C. M., and Vohralik, P. F.: Three-dimensional simulations of long-lived tracers using winds from MACCM2, J. Geophys. Res., 102, 21493–21513, doi:10.1029/97JD00793, 1997.

- Worden, J., Kulawik, S. S., Shepard, M., Clough, S., Worden, H., Bowman, K., and Goldman, A.: Predicted errors of Tropospheric Emission Spectrometer nadir retrievals from spectral window selection, J. Geophys. Res., 109, D09308, doi:10.1029/2004JD004522, 2004.
- Worden, H. M., Bowman, K. W., Worden, J. R., Eldering, A., and Beer, R.: Satellite measurements of the clear-sky greenhouse effect from tropospheric ozone, Nat. Geosci., 1, 305–308, 2008.
- Worden, J., Dylan, B., Jones, A., Liu, J., Parrington, M., Bowman, K., Stajner, I., Beer, R., Jiang, J., Thouret, V., Kulawik, S., Li, J. F., Verma, S., and Worden, H.: Observed vertical distribution of tropospheric ozone during the Asian summertime monsoon, J. Geophys. Res., 114, D13304, doi:10.1029/2008JD010560, 2009.
- Zhang, Y., Takahashi, M., and Guo, L.: Analysis of the East Asian Subtropical Westerly Jet Simulated by CCSR/NIES/FRCGC Coupled Climate System Model, J. Meteoro. Soc. Jpn., 86, 257–278, 2008.